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Visualizing flow vortices inside a single levitated drop

A. Amar [a,*], E. Groß-Hardt [b], A.A. Khrapitchev [a], S. Stapf [a], A. Pfennig [b], B. Blümich [a]

[a] *Institute of Technical and Macromolecular Chemistry, ITMC, RWTH Aachen, Worringerweg 1, D-52074 Aachen, Germany*
[b] *Department of Chemical Engineering/Thermal Unit Operations, TVT, RWTH Aachen, Wüllnerstr. 5, D-52062 Aachen, Germany*

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Abstract

The internal flow dynamics in single liquid drops, kept in place through levitation by a counterflowing continuous fluid phase in a suitably designed glass cell, is investigated by PFG NMR techniques. The positional stability of the drops was confirmed from series of one-dimensional profiles and was found to be below the spatial resolution of the experiment. Velocity distribution functions (propagators) along all three coordinates were obtained and demonstrated the long-time stability of the internal dynamics in terms of the velocity magnitudes occurring in the systems. Finally, velocity imaging was applied to visualize the internal vortex patterns in the drops either as projections onto different planes or within thin slices of selected orientations. Two different fluid systems were investigated in order to cover the principal cases of rigid and mobile interfaces. Different fast velocity imaging techniques were employed for monitoring the vastly differing velocity ranges of both cases, and the high sensitivity of the internal three-dimensional motion to the cell geometry is demonstrated.

Keywords: Velocity imaging; Fluid dynamics; PFG NMR; Propagator; Vortices

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1. Introduction

Liquid–liquid extraction processes are of widespread use in chemical engineering and have their most important application in cleaning procedures where contaminants in a bulk, valuable fluid component (donator phase) are being removed by bringing it into contact with a second, disperse phase (acceptor phase). Ideally, donator and acceptor phase are immiscible, while the contaminant (transfer phase) is soluble in both fluids. To provide maximum transfer within a given amount of time, a large concentration difference of the contaminant and a large interface area between the two main phases are desired. This is often realized by dispersing the acceptor phase into a swarm of droplets and allowing it to pass through the continuous phase exploiting the density differences between phases.

It is a well-known fact that the efficiency of mass-transfer between the two phases is determined by convective transport made possible through circulation occurring both inside and outside of the droplets. Mass-transfer can be, in fact, substantially faster than would be expected from pure diffusive transport across the drop interface. Mass-transfer rates are underestimated by orders of magnitude by the analytical solution of Kronig and Brink [1], but also by 2D-axisymmetric CFD simulations for non-deformable droplets with an ideally mobile interfacial region, which do not make use of approximated solutions of the Navier Stokes equations [2,3]. Modelling mass-transfer, however, depends on a precise knowledge of the fluid dynamics inside the drop, which in turn can be understood theoretically only by taking into account sufficiently detailed models of the boundary layer properties. The single-droplet behaviour, which needs to be understood as a basis for the extraction-column design, is determined by mass-transfer and sedimentation, which take place

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[*] Corresponding author. Fax: +49 241 80 22185.
E-mail address: aamar@mc.rwth-aachen.de (A. Amar).

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simultaneously and influence each other. Although in the past, several theoretical, numerical, and experimental investigations on single droplets have been carried out, sedimentation velocities and mass-transfer rates cannot be predicted a priori; experimental data can only be matched by additional empirical parameters. The only experimental evidence for fluid dynamics is usually delivered from integral measurements of the mass-transfer in an extraction column or in single-drop cells [4,5]. Although particle tracer methods have been used to visualize the flow pattern in drops directly [6–8], these are limited in their applicability with respect to resolution and dimensionality, frequently monitoring only motion in suitable sections within the drop. Furthermore, they represent an invasive technique which can compromise the validity of the results derived about the fluid flow field. For instance, it is known that the fluid dynamics of the drop can be very sensitive to small concentrations of impurities in the system which tend to accumulate at the interface.

Pulsed field gradient (PFG) NMR appears to be an exceptionally suitable technique for non-invasively monitoring the drop's internal fluid dynamics and its change with time. In recent literature, the versatility of velocity encoded imaging and its applicability to model systems and problems from the field of chemical engineering were demonstrated. Methods based on conventional imaging are often prohibitively slow to achieve sufficient spatial resolution in a reasonable experimental time which is required to monitor processes that are potentially instationary (see the compilation about flow NMR in [9]). Therefore, several attempts were made to combine multi-pulse and/or multi-acquisition imaging techniques with velocity encoding modules. While a long lifetime of the signal and a comparatively slow motion favours repeated refocusing as achieved by turbo spin echo/RARE [10] or EPI [11], gradient-recalled echo techniques following small flip angles as in FLASH [12] appear more appropriate if the signal lifetime is short. Sederman et al. [13] have demonstrated the feasibility of fast imaging in combination with velocity encoding to visualize transient phenomena and to determine spatially resolved velocity autocorrelation functions. However, their spatial resolution was optimized to fit the comparatively large size of the sample under study. Several more recent approaches to systems of different requirements have tackled the problem of combining high-resolution velocity measurements with fast imaging techniques [14–21]. In [22], velocities inside a small (3.5 mm) falling water drop were visualized, but with rather long experimental times due to the need to accumulate the desired information from a sufficient number of free falling individual drops.

In the present study, levitated single drops inside a continuous, liquid phase were investigated. They were kept in place by adjusting the counter current of the continuous phase in a suitably shaped device that is located inside the magnet bore; this apparatus is briefly introduced in Section 2.1. The drops of typically 2–4 mm in diameter held in this set-up had to be imaged with sufficient spatial resolution. The internal dynamics of the drops can generally be divided into different regimes. While small droplets sediment like rigid spheres, larger droplets feature pronounced internal dynamics; with knowing the properties of the substances the diameter of transition between these limiting cases can be estimated by the purely empiric 'Bond criterion.' A better indicator are measured sedimentation velocities in comparison to the CFD-simulations of the limiting cases with a mobile or a rigid interfacial region. It is one purpose of this study to discuss these limiting cases, which requires the determination of either very small or very large velocities in an otherwise identical geometry. The fact that the drops did not move as a whole allowed the application of multiple acquisition techniques, being compromised only by the need to allow full relaxation of the spin system by introducing sufficient delays in between signal encodings, contrary to the conditions in [22].

Empirical estimates do exist for predicting the dynamic properties of drops, but do require ideal systems of sufficient purity. In reality, the transition between rigid and mobile interfaces is smooth and the precise properties of the boundary are not known in detail. To discuss clearly distinct cases, two types of liquids were used as the disperse phase, namely a low-molecular weight silicone oil (octamethylcyclotetrasiloxane, OMCTS) and toluene. Small (2 mm) OMCTS drops and large (4 mm) toluene drops were generated in order to compare the limiting conditions of rigid and mobile drop interfaces. The varying ranges of velocities encountered with these systems made necessary the application of two different PFG techniques. In the experimental part of this paper, the results of stability tests by means of spin-density imaging of the drop are presented, followed by a discussion of the velocity distribution in terms of statistical measures as well as velocity maps in different spatial directions.

2. Experimental

2.1. Set-up of the levitating device

The foremost requirement for measuring internal dynamics in levitated drops is the generation of a drop being stable in position and shape for long periods of time, at least on scales below the desired resolution in the spatial and velocity dimensions. Furthermore, reproducibility of drop generation and position are needed for measurements of series of drops. A set of conical glass cells was manufactured with different geometrical parameters and degrees of skewness; some cells were